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Abstract

We present changes in volume, immersion density, and tensile properties observed from accelerated aged plutonium alloys. Accelerated alloys (or spiked alloys) are plutonium alloys enriched with approximately 7.5 weight percent of the faster-decaying ^{238}Pu to accelerate the aging process by approximately 17 times the rate of unaged weapons-grade plutonium. After sixty equivalent years of aging on spiked alloys, the dilatometry shows the samples at 35°C have swelled in volume by 0.15 to 0.17 % and now exhibit a near linear volume increase due to helium in-growth. The immersion density of spiked alloys shows a decrease in density, similar normalized volumetric changes (expansion) for spiked alloys. Tensile tests show increasing yield and engineering ultimate strength as spiked alloys are aged.

INTRODUCTION

Plutonium, because of its radioactive nature, ages from the "inside out" by means of self-irradiation damage and thus produces Frenkel-type defects (vacancies and self-interstitial atoms) and defect clusters [1, 2]. The self-radiation damage in Plutonium-239 occurs mainly by alpha-particle decay, where most of the damage comes from the ^{235}U recoil nucleus. The defects resulting from the residual lattice damage and helium in-growth could result in microstructural and physical property changes. Because these self-irradiation effects would normally require decades to measure, a fraction (7.5 wt%) of more active isotope ^{238}Pu is added to the reference plutonium alloy thus accelerating the aging process by approximately 17 times. By monitoring the properties of the Pu-238 spiked alloy over a period of about 3.5 years, the properties of plutonium can be projected for periods up to about 60 years. This paper presents density, volume, and tensile changes observed from accelerated aged plutonium alloys.

EXPERIMENT

Each dilatometer unit consists of a small vacuum controlled-atmosphere sample chamber fitted with three linear variable differential transducers (LVDTs). The schematic and detailed operations of the dilatometer system are presented elsewhere [3, 4], so only a brief description is provided here. An LVDT measures minute changes, 0.1 micron or less, in the position of a push-rod by monitoring changes in the inductance of a detector coil. In the current design, the

detector coil is placed outside of the sample chamber. Two different lengths (2 and 3 cm) of alloy specimens are used to differentiate between surface oxidation and volumetric swelling in the materials. These alloys have nominal gallium concentration of 0.5 weight percentage. These specimens are placed in the copper well located inside the dilatometer system at 35°C storage temperatures. A reference low thermal expansion glass (Zerodur) is also placed in the copper well to monitor the stability of the dilatometry system.

The immersion density equipment closely matches a design used by Bowman et al. [5] and uses about 200 ml of Fluorinert Electronic Liquid FC-43 as the immersion fluid. Prior to use, the system is calibrated using NIST glass (SRM 1827A). Because the ^{238}Pu-spiked alloys generate heat, a test sample is left overnight in the immersion bath to allow the temperature of the bath to stabilize and the measurements to be reproducible. A correction needs to be applied to the measured density to compensate for the heat generated by the ^{238}Pu-spiked sample. Details of density correction are described elsewhere [4].

Each tensile test specimen was dimensionally inspected for the gage diameter and its length (GL), and loaded into a specially designed fixture for the tensile test. With a 0.24 inch GL extensiometer for the strain measurement, all testing was performed at crosshead speed of 0.05 inch/min, so that the ultimate strain rate was about 3.5×10^{-3}/sec. Aluminum specimens of known tensile strength were tested to verify the equipment integrity before and after each test specimen. The load and displacement data was recorded on a computer using the Instron Series IX software package in conjunction with an Instron Model 4444 test machine. Data was recorded from preloading until failure. Plutonium specimens for the tensile test have nominal gallium concentration of 1 weight percentage.

DISCUSSION

The volume change (ΔV) normalized with the initial volume (V_o) of spiked alloys is shown in Figure 1. The time is represented as an equivalent time (in year) obtained by multiplying the measurement time by the accelerating factor (e.g. an average factor of 17). To calculate this factor, the α-decay activities of the ^{238}Pu-enriched alloy and the reference alloy are determined using the concentration of isotopes in each alloy. Then the activity of the enriched alloy is normalized to that of the reference alloy to obtain the factor. This accelerating factor will decrease as the material ages due primarily to decreasing concentration of ^{238}Pu in the specimen. Since the volume expansion due to self-irradiation damage is assumed isotropic in the bulk of material and is small compared to the total volume, the $\Delta V/V_o$ of the specimen can be obtained with the approximate relation $\Delta V/V_o \cong 3\ \Delta L/L_o$ where $\Delta L/L_o$ represents the measured specimen length change (ΔL) normalized with the initial length (L_o). As plotted in Figure 1, all the spiked alloys have increased in volume significantly. During the early stage of measurement, samples at 35°C storage temperature increased in volume as a result of self-irradiation and follow the inverse exponential-type of expansion on dose or time during initial stage of aging. This behavior has been attributed to the lattice damaged caused by the radiation damage. After the initial expansion, the volume change exhibits a

significantly lower rate of increase and a near linear expansion behavior attributed to the helium in-growth mechanism.

The curves in Fig. 1 are quite accurately represented by the combination of exponential and linear growth equations of the form [4]

$$\Delta V/V_o \cong A\,[1\text{-}\exp(-Bt)] + Ct \qquad (1)$$

where A, B, and C are constants and t is the time in years. The He/vacancy association ratios are calculated using the slope (C). This ratio describes the volume expansion induced by the formation of the helium bubbles in Pu metal. The average ratio extracted from the curve fit is approximately 2.5.

Figure 2 shows results of immersion density measurement on the reference (identical composition to spiked alloys with the exception of the ^{238}Pu) and ^{238}Pu-spiked alloys. The ages of reference samples range from 0.2 to 21 years. Large error bars in density data originates primarily from the effects of convection currents (heating) in the immersion fluid generated by the alpha decay of plutonium. The initial density value for the dilatometry data is set to 15.795 g/cc, which is the measured initial reference alloy density, to compare to density changes of the reference alloys. Following the initial transient reduction in density, the rate of density reduction becomes reduced as observed from the dilatometry.

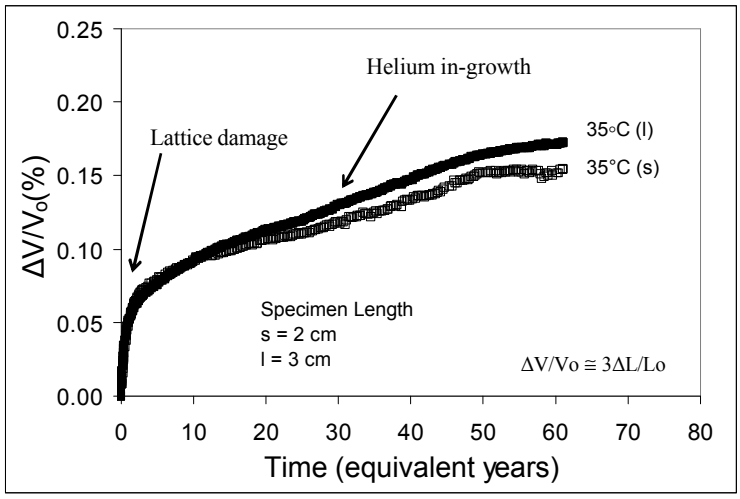

Figure 1. The normalized volume changes for spiked alloys tested at 35°C. 2 and 3 cm length samples are described as “s” and “l”.

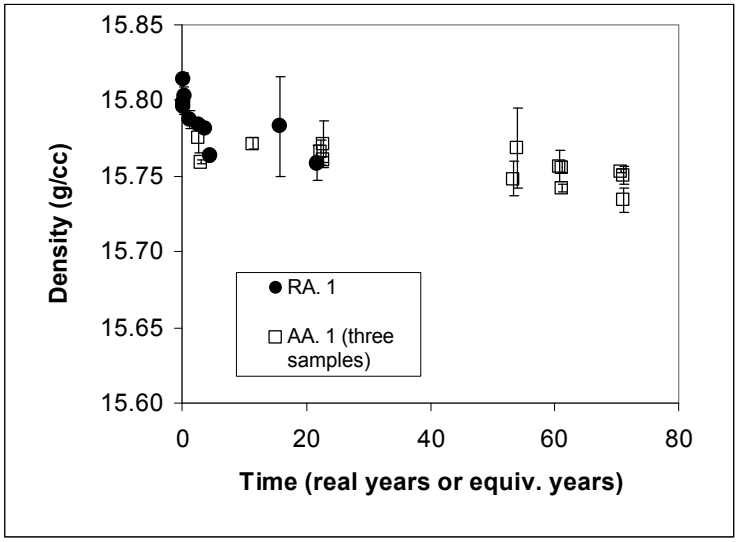

Figure 2. Both reference alloys (RA.1) and spiked alloys (AA.1) show decreasing density as alloys are aged.

Figure 3 shows the stress-strain curves of spiked alloys at 3, 40, and 70 equivalent years, respectively. The yield strength and the engineering ultimate strength of the spiked alloy increase significantly with age. The Young's modulus is very consistent at 40 GPa between tested samples irrespective of age.

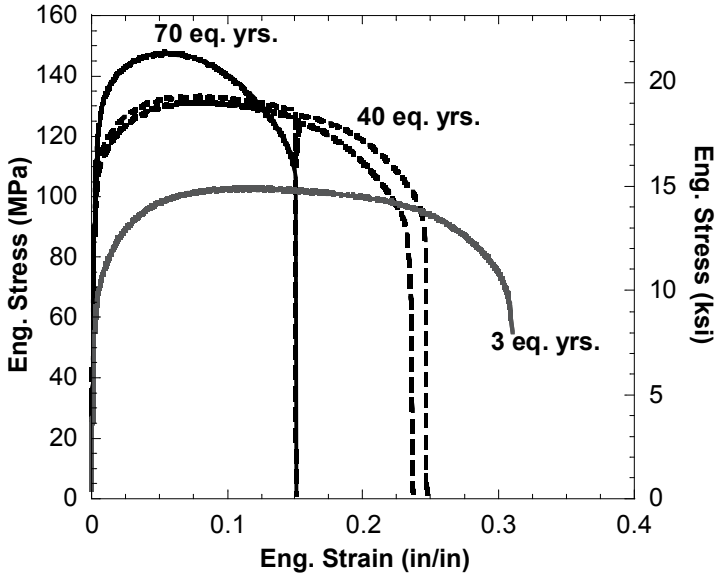

Figure 3. Engineering stress versus Engineering strain for spiked alloys at 4, 40, and 70 equivalent years.

CONCLUSIONS

Dilatometry, immersion density, and tensile measurements are underway on ^{238}Pu-enriched alloys. Current measurements show the volume expansion, density reduction, and strength increase from the accumulation of residual lattice damage and

helium in-growth. By modeling the volume swelling behavior, we estimate the average He/vacancy ratio to be about 2.5 indicating that the helium bubble pressure is approximately in equilibrium with the surface tension of the bubble formed in plutonium alloy.

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